

polyatomic ions comprising a plurality of sub-populations of ions, all the ions of each of said sub-populations having the same charge state number, said same charge state number differing from the charge state numbers of the ions in the other sub-populations of said plurality of subpopulations, said plurality of sub-populations comprising one sub-population for each value of charge state number beginning with a smallest value not less than three and extending to a largest value not less than five.

*cont'd*  
Claim 68<sup>2</sup> (amended). The composition of matter of claim 67 in which said smallest value of charge state number is not less than five and said largest value is not less than seven.

Claim 69<sup>3</sup> (amended). The composition of matter of claim 67 in which said smallest value of charge state number is not less than seven and said largest value is not less than ten.

Claim 70<sup>4</sup>. The composition of matter of claim 67 in which said population of multiply charged polyatomic ions is formed by:

dispersing a solution containing said one or more distinct polyatomic parent molecular species into a bath gas as charged droplets, said dispersing taking place in the presence of an electric field; and allowing the solvent of said solution to evaporate from said charged droplets until at least some molecules of said distinct polyatomic parent molecular species become dispersed in said bath gas as said multiply charged polyatomic ions.

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Claim 7<sub>1</sub> (amended) The composition of matter of claim 6<sub>7</sub> in which said polyatomic parent molecular species is selected from a class of compounds known as biopolymers.

6  
Claim 7<sub>2</sub> (amended) The composition of matter of claim 6<sub>7</sub> in which said distinct polyatomic parent molecular species is not a synthetic polymer, such as a poly (ethylene glycol), having less than four different constituent elemental species.

7  
Claim 7<sub>3</sub> (amended) The composition of matter of claim 6<sub>7</sub> such that a mass analysis of said multiply charged polyatomic ions in said population of multiply charged polyatomic ions comprising a plurality of sub-populations, produces a set of values for the mass/charge ratios of the multiply charged polyatomic ions in said population of ions, said set of values of mass/charge ratios providing a basis for calculating a value of molecular weight for the said polyatomic parent molecular species from which said population of multiply charged polyatomic ions is formed.

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Claim 7<sub>4</sub> (amended) The composition of matter as claimed in claim 6<sub>7</sub> in which said distinct polyatomic parent molecular species has a molecular weight not less than 5000.

9  
Claim 7<sub>5</sub> (amended) A composition of matter comprising one or more populations of multiply charged polyatomic ions derived from a sample comprising at least one polyatomic parent molecular species, the number of charges on each ion defining said ion's charge state number, said population of multiply charged polyatomic ions formed

from said at least one polyatomic parent molecular species comprising a plurality of sub-populations, the ions of each sub-population having the same charge state number, said charge state number differing by one from the next largest and the next smallest values of charge state number found in the other sub-populations of said plurality, the ions of each of said sub-populations having a value of said charge state number that is not less than five, said composition of matter being formed by:

dispersing a solution of said sample containing said at least one polyatomic parent molecular species into a bath gas as charged droplets, said dispersing taking place in the presence of an electric field; and

*cont'd*  
*El*

allowing the solvent of said solution to evaporate from said charged droplets until at least some molecules of said polyatomic parent molecular species become dispersed in said bath gas as said multiply charged polyatomic ions.

10 9  
Claim 76. The composition of matter of claim 75 in which the charge state number of the ions in each of said sub-populations of said plurality of sub-populations is at least seven.

11 9  
Claim 77. The composition of matter of claim 75 in which all molecules of said at least one of said polyatomic parent molecular species have substantially the same molecular weight.

*12* 9  
Claim ~~78~~ 9. The composition of matter of claim ~~75~~ in which said single polyatomic parent molecular species is selected from the class of compounds known as biopolymers.

*13* 9  
Claim ~~79~~ (amended). The composition of matter of claim ~~75~~ such that when a mass spectrum is generated from said population of multiply charged polyatomic ions, an analysis of the mass/charge values of the ions giving rise to each of said peaks in said sequence of peaks in said mass spectrum leads to a value of the molecular weight of said single polyatomic parent molecular species.

*Con't*

*15* 9  
Claim ~~80~~ 9. The composition of matter of claim ~~75~~ in which said single polyatomic parent molecular species has a molecular weight of not less than about 5000.

*14* 13  
Claim ~~81~~ 13. The composition of matter of claim ~~75~~ in which said single polyatomic parent molecular species has a molecular weight not less than 5000.

*16* 13  
Claim ~~82~~ (amended). A composition of matter comprising one or more distinct populations of multiply charged polyatomic ions generated from a sample comprising one or more distinct polyatomic parent molecular species, the number of charges on each ion defining the ion's charge state number, each of said populations of polyatomic ions comprising a plurality of sub-populations, each of said plurality of sub-populations being comprised of ions formed from one of said distinct polyatomic parent molecular species and having the same charge state

number, there being at least one of said populations of multiply charged polyatomic ions that comprises one of said sub-populations for each value of charge state number beginning with a smallest value of three and extending to a largest value not less than five.

*17*  
Claim 83 (amended). The composition of matter of claim 82 in which said smallest value of charge state number is not less than five and said largest value of charge state number is not less than seven.

*18*  
Claim 84 (amended). The composition of matter of claim 82 in which said smallest value of charge state number is not less than seven and said largest value of charge state number is not less than ten.

*19*  
Claim 85. The composition of matter of claim 82 in which all molecules of each of said distinct polyatomic parent molecular species have substantially the same molecular weight.

*20*  
Claim 86. The composition of matter in claim 82 in which at least one of said distinct polyatomic parent molecular species is selected from a class of compounds known as biopolymers.

*21*  
Claim 87. The composition of matter in claim 82 in which at least one of said distinct polyatomic parent molecular species is selected from the group comprising proteins, peptides, polypeptides, carbohydrates, oligonucleotides and glycoproteins.

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Claim 28 (amended). The composition of matter of claim 82 in which at least one of said distinct polyatomic parent molecular species is not a synthetic polymer, such as a poly (ethylene glycol), having less than four different constituent elemental species.

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Claim 29 (amended). The composition of matter of claim 82 in which at least one of said distinct polyatomic parent molecular species has a molecular weight not less than about 5000.

CONT  
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Claim 20 (amended). A composition of matter comprising one or more distinct populations of multiply charged polyatomic ions generated from a sample comprising one or more distinct polyatomic parent molecular species, the number of charges on each ion defining its charge state number, each of said populations of multiply charged polyatomic ions comprising ions formed from one of said distinct polyatomic molecular species and being comprised of a plurality of sub-populations, the ions of each of said sub-populations having the same charge state number, there being one of said sub-populations for each value of said charge state number beginning with a smallest value not less than three and extending to a largest value not less than five.

11.25

Claim 21 (amended). The composition of matter of claim 20 in which said smallest value of charge state number is at least five and said largest value is not less than seven.

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*26* *24*  
Claim ~~92~~. The composition of matter of claim ~~90~~ in which at least one of said distinct polyatomic parent molecular species is selected from a class of compounds known as biopolymers.

*27* *24*  
Claim ~~93~~. The composition of matter of claim ~~90~~ in which at least one of said distinct polyatomic parent molecular species is selected from the group comprising proteins, peptides polypeptides, carbohydrates, oligonucleotides and glycoproteins.

*28* *24*  
Claim ~~94~~ (amended). The composition of matter of claim ~~90~~ in which at least one of said distinct polyatomic parent molecular species is not selected from the group comprising synthetic polymers having less than four different constituent elemental species, said group comprising poly (ethylene glycol)s.

*29* *24*  
Claim ~~95~~ (amended). The composition of matter of claim ~~90~~ in which at least one of said distinct polyatomic parent molecular species has a molecular weight not less than 5000.

*30* *24*  
Claim ~~96~~ (amended). The composition of matter of claim ~~90~~ in which at least one of said distinct polyatomic parent molecular species has a molecular weight not less than 7000.

*31*  
Claim ~~97~~ (amended). A composition of matter comprising one or more distinct populations of multiply charged polyatomic ions generated from a sample comprising one or more distinct polyatomic parent molecular species, the number of charges on each ion defining the ion's

charge state number, each of said populations of multiply charged polyatomic ions comprising ions formed from one of said distinct polyatomic parent molecular species in said sample, at least one of said populations of multiply charged polyatomic ions being comprised of a plurality of sub-populations, the ions of each of said sub-populations having the same value of charge state number, that value being different from the values of charge state number in all the other sub-populations of ions in said plurality of sub-populations, the smallest value of charge state number of the ions in said plurality of sub-populations being not less than three, said composition of matter being formed by:

cont'd

dispersing a solution containing said one or more distinct polyatomic parent molecular species into a bath gas as charged droplets, said dispersing taking place in the presence of an electric field.

allowing the solvent of said solution to evaporate from said charged droplets until at least some molecules of said distinct polyatomic parent molecular species become dispersed in said bath gas as said multiply charged ions.

7 32 31  
Claim 98 (amended). The composition of matter of claim 97 in which said smallest value of charge state number is not less than five.

33 31  
Claim 99 (amended). The composition of matter of claim 97 in which said smallest value of charge state number is not less than seven.

34

Claim 100-(amended). The composition of matter of claim 97 in which all molecules of any one of said distinct polyatomic parent molecular species have substantially the same molecular weight.

35

Claim 101. The composition of matter of claim 97 in which all molecules of at least one of said distinct polyatomic parent molecular species have the same chemical formula.

36

Claim 102 (amended). The composition of matter of claim 97 in which all molecules of each of said distinct polyatomic parent molecular species are chemically indistinguishable.

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Claim 103. The composition of matter of claim 97 in which at least one of said distinct polyatomic parent molecular species is selected from the class of compounds known as biopolymers.

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Claim 104 (amended). The composition of matter of claim 97 in which at least one of said distinct polyatomic parent molecular species is not selected from the group of synthetic polymers having less than four different constituent elemental species, said group comprising poly (ethylene glycol)s.

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Claim 105 (amended). The composition of matter of claim 97 in which at least one of said distinct polyatomic parent molecular species has a molecular weight not less than about 5000.

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Claim 106. The composition of matter of claim 97 in which said bath gas is heated.

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Claim 107. The composition of matter of claim 97 in which said bath gas flows in a direction substantially counter current to the direction in which said charged droplets drift in said electric field.

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Claim 108. The composition of matter of claim 97 in which said bath gas is at approximately atmospheric pressure.

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Claim 109. A composition of matter comprising one or more populations of multiply charged polyatomic ions generated from a sample comprising one or more distinct polyatomic parent molecular species, the number of charges on each ion defining the ion's charge state number, each of said populations of multiply charged polyatomic ions comprising ions formed from one of said one or more distinct polyatomic parent molecular species, at least one of said populations of ions comprising a plurality of sub-populations of ions, all the ions in each sub-population having the same charge state number, said at least one of said populations comprising one such sub-population for each possible value of charge state number beginning with a smallest value not less than three and extending to a largest value not less than five, said composition of matter being useful in the determination of a value of molecular weight for one or more of said distinct polyatomic parent molecular species, said determination of molecular weight being achieved by means of a mass analysis of ions from said one or more populations of ions and a calculation of the molecular weight

values of said one or more polyatomic parent molecular species from the values of mass/charge (m/z) obtained by said mass analysis for the ions in said one or more populations of polyatomic ions.

*44*  
Claim ~~110~~ (amended). The composition of matter of claim ~~109~~ in which said smallest value of charge state number is not less than five and said largest value of charge state number is not less than seven.

*45*  
Claim ~~111~~ (amended). The composition of matter of claim ~~109~~ in which said smallest value of charge state number is not less than seven and said largest value of charge state number is not less than ten.

*46*  
Claim ~~112~~ (amended). The composition of matter of claim ~~109~~ in which all molecules of any particular one of said distinct polyatomic parent molecular species have substantially the same molecular weight.

*47*  
Claim ~~113~~. The composition of matter of claim ~~109~~ in which at least one of said distinct polyatomic parent molecular species is selected from a class of compounds known as biopolymers.

*48*  
Claim ~~114~~. The composition of matter of claim ~~109~~ in which at least one of said distinct polyatomic parent molecular species is selected from the group comprising proteins, peptides, polypeptides, carbohydrates, oligonucleotides and glycoproteins.

*49*  
Claim ~~115~~ (amended). The composition of matter of claim ~~109~~ in which at least one of said distinct polyatomic parent molelcular species is

not selected from the group of synthetic polymers having less than four different distinct elemental constituent species, said group comprising poly (ethylene glycol)s.

*50* *43*  
Claim ~~116~~ (amended). The composition of matter of claim ~~109~~ in which at least one of said distinct polyatomic parent molecular species has a molecular weight not less than 5000.

*51*  
Claim ~~117~~ (amended). The composition of matter comprising one or more populations of multiply charged polyatomic ions generated from a sample comprising one or more distinct polyatomic parent molecular species, the number of charges on each ion defining the ion's charge state number, each of said populations comprising ions formed from one of said one or more distinct polyatomic parent molecular species, at least one of said populations of multiply charged polyatomic ions comprising a plurality of sub-populations of ions, all the ions in each sub-population having the same charge state number, said same charge state number differing from the charge state numbers of the ion in the other sub-populations of said population, said charge state number having a value of at least five for all the ions in said at least one of said populations of multiply charged polyatomic ions, said composition of matter being useful for determining the molecular weight of one or more of said distinct polyatomic parent molecular species, said determination of the molecular weight being achieved by a mass analysis of the ions in said one or more populations of multiply charged polyatomic ions together with a calculation of the said molecular weight of said one or more polyatomic parent molecular species

from the values of mass/charge (m/z) obtained by mass analysis of ions in said one or more populations of multiply charged polyatomic ions.

*52* *51*  
Claim ~~118~~ (amended). The composition of matter of claim ~~117~~ in which every ion in said at least one of said populations of multiply charged polyatomic ions has a charge state number not less than seven.

*53* *51*  
Claim ~~119~~. The composition of matter of claim ~~117~~ in which at least one of said distinct polyatomic parent molecular species is selected from a class of compounds known as biopolymers.

*54* *51*  
Claim ~~120~~. The composition of matter of claim ~~117~~ in which at least one of said distinct polyatomic parent molecular species is selected from the group comprising proteins, peptides, polypeptides, carbohydrates, oligonucleotides and glycoproteins.

*55* *51*  
Claim ~~121~~ (amended). The composition of matter of claim ~~117~~ in which at least one of said distinct polyatomic parent molecular species is not selected from the group comprising poly (ethylene glycol)s.

*56* *51*  
Claim ~~122~~ (amended). The composition of matter of claim ~~117~~ in which at least one of said distinct polyatomic parent molecular species has a molecular weight not less than 5000.

*57*  
Claim ~~123~~ (amended). A composition of matter comprising one or more distinct populations of multiply charged polyatomic ions generated from a sample comprising one or more distinct polyatomic parent mole-

*cont'd*

molecular species, the number of charges on each ion defining the ion's charge state number, each of said multiply charged polyatomic ions in any one of said one or more distinct populations having been formed from one of said distinct polyatomic parent molecular species in said sample, at least one of said distinct populations of multiply charged polyatomic ions comprising a plurality of sub-populations of ions, all the ions in each sub-population of said plurality of sub-populations having the same charge state number, said same charge state number differing from the charge state numbers of the ions in the other sub-populations of said plurality of sub-populations, said plurality of sub-populations comprising one such sub-population for each possible value of charge state number beginning with a smallest value not less than three and extending to a largest value not less than five, said composition of matter being formed by:--

dispersing a solution containing said polyatomic parent molecular species into a bath gas as charged droplets, said dispersing taking place in the presence of an electric field;

allowing the solvent of said solution to evaporate from said charged droplets until at least some molecules of said distinct polyatomic parent molecular species become dispersed in said bath gas as said multiply charged polyatomic ions;

said composition of matter having the property that the molecular weight of each of said distinct polyatomic parent molecular species in said sample can be calculated from the mass/charge (m/z) values of the multiply charged polyatomic ions produced from that species.

*58*  
Claim ~~124~~ (amended). The composition of matter of claim ~~123~~ in which said smallest value of charge state number is not less than five and said largest value is not less than seven.

*59*  
Claim ~~125~~ (amended). The composition of matter of claim ~~123~~ in which said smallest value of charge state number is not less than seven and said largest value is not less than ten.

*60*  
Claim ~~126~~ (amended). The composition of matter of claim ~~123~~ in which all molecules of any particular one of said distinct polyatomic parent molecular species have substantially the same molecular weight.

*61*  
Claim ~~127~~. The composition of matter of claim ~~123~~ in which at least one of said distinct polyatomic parent molecular species is selected from the class of compounds known as biopolymers.

*62*  
Claim ~~128~~. The composition of matter of claim ~~123~~ in which at least one of said distinct polyatomic parent molecular species is selected from the group comprising proteins, peptides, polypeptides, carbohydrates, oligonucleotides and glycoproteins.

*63*  
Claim ~~129~~ (amended). The composition of matter of claim ~~123~~ in which at least one of said distinct polyatomic parent molecular species is not selected from the group of synthetic polymers comprising less than four different constituent elemental species, said group comprising poly (ethylene glycol)s.

64 51  
Claim ~~120~~ (amended). The composition of matter of claim ~~123~~ in which at least one of said distinct polyatomic species has a molecular weight not less than 5000.

65 51  
Claim ~~124~~. The composition of matter of claim ~~123~~ in which said bath gas is heated.

66 51  
Claim ~~122~~ (amended). The composition of matter of claim ~~123~~ in which said bath gas flows in a direction substantially counter current to the direction of drift of said charged droplets in said electric field.

67 51  
Claim ~~123~~. The composition of matter of claim ~~123~~ in which said bath gas is at approximately atmospheric pressure.

68 51  
Claim ~~124~~ (amended). A composition of matter that by mass analysis of its component ions is found to comprise one or more distinct populations of multiply charged polyatomic ions, the number of charges on each ion defining the ion's charge state number, each of said distinct populations of multiply charged polyatomic ions comprising ions having been formed from a polyatomic parent molecular species, at least one of said distinct populations of multiply charged polyatomic ions comprising a plurality of sub-populations of ions, all the ions in each sub-population having the same charge state number, said charge state number differing from the charge state number of the other sub-populations in said plurality of sub-populations, said plurality of sub-populations comprising one such sub-population for each possible value of charge state number beginning with a smallest value not less

than three and extending to a largest value not less than five, said composition of matter being formed by:

dispersing a solution containing one or more polyatomic molecular species into a bath gas as charged droplets, said dispersing taking place in the presence of an electric field;

allowing the solvent of said solution to evaporate from said charged droplets until at least some molecules of said polyatomic parent molecular species become dispersed in said bath gas as said multiply charged polyatomic ions;

said mass analysis being carried out on a portion of said multiply charged polyatomic ions in said bath gas that is introduced into a vacuum system containing a mass analyzer.

*69*  
Claim ~~135~~ (amended). The composition of matter of claim ~~134~~ in which said smallest value of charge state number is not less than five and said largest value is not less than seven.

*70*  
Claim ~~136~~ (amended). The composition of matter of claim ~~134~~ in which said smallest value of charge state number is not less than seven and said largest value is not less than ten.

*71*  
Claim ~~137~~ (amended). The composition of matter of claim ~~134~~ in which all molecules of any one of said distinct polyatomic parent molecular species in said solution have the same chemical identity.

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Claim 138. The composition of matter of claim 134 in which at least one of said distinct polyatomic parent molecular species in said solution is selected from a class of compounds known as biopolymers.

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Claim 139 (amended). The composition of matter of claim 134 in which at least one of said distinct polyatomic parent molecular species in said solution has a molecular weight not less than 5000.

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Claim 140 (amended). A composition of matter derived from a sample comprising one or more distinct polyatomic parent molecular species, all molecules of each of said distinct polyatomic parent molecular species having substantially the same molecular weight and chemical identity, said composition of matter comprising one or more distinct populations of polyatomic ions, at least one of said distinct populations of ions comprising multiply charged ions formed from one of said one or more distinct polyatomic parent molecular species in said sample, the number of charges on each ion defining the charge state number of that ion, each of said populations of multiply charged ions having the property that when its ions are mass analyzed they give rise to a mass spectrum comprising a multiplicity of peaks, said multiplicity of peaks comprising at least one coherent sequence of peaks, the ions of each peak in said coherent sequence having the same charge state number, said charge state number being greater than unity and differing by one unit from the charge state numbers of the ions of each immediately adjacent peak in said coherent sequence, said coherent sequence of peaks comprising one peak for each different

value of charge state number beginning with a smallest value not less than three and extending to a largest value not less than five.

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Claim 144 (amended). The composition of matter of claim 140 in which said smallest value of charge state number is not less than five and said largest value of charge state number is not less than seven.

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Claim 142. The composition of matter of claim 140 in which at least one of said distinct polyatomic parent molecular species is selected from a class of compounds known as biopolymers.

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Claim 143. The composition of matter of claim 140 in which at least one of said distinct polyatomic parent molecular species is not selected from the group comprising poly (ethylene glycol)s.

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Claim 144 (amended). The composition of matter of claim 140 in which at least one of said distinct polyatomic parent molecular species has a molecular weight not less than about 5000.

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Claim 145 (amended). The composition of matter of claim 140 in which at least one of said distinct polyatomic parent molecular species has a molecular weight not less than 7000.

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Claim 146 (amended). A composition of matter comprising one or more distinct populations of multiply charged polyatomic ions, each of said multiply charged polyatomic ions in said distinct populations being characterized by the symbol  $x_i$ , the numerical value of  $x_i$  being

the m/z value of said one of said multiply charged ions such that  
 $xi = Mr/i + ma$  wherein Mr is the molecular weight of a distinct parent polyatomic molecular species from which all of said multiply charged polyatomic ions in said distinct population of multiply charged polyatomic ions are derived, i is an integer equal to the number of adduct charges attached to a molecule of said distinct polyatomic parent molecular species to form one of said multiply charged polyatomic ions, ma is the effective average mass (which can sometimes be negative) of each said adduct charge, each of said distinct populations of multiply charged polyatomic ions comprising a plurality of sub-populations, the ions of each sub-population having the same values for i, ma and Mr and therefore the same value of xi, said distinct populations of ions including at least one population in which all the ions have values of i greater than two.

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81 86  
Claim 147 (amended). The composition of matter of claim 146 in which said one or more distinct populations of multiply charged polyatomic ions include at least one population in which all the ions have values of i greater than three.

82 80  
Claim 148 (amended). The composition of matter of claim 146 in which said one or more distinct populations of multiply charged polyatomic ions include at least one population in which all the ions have values of i greater than five.

83 80  
Claim 149 (amended). The composition of matter of claim 146 in which at least one of said polyatomic parent molecular species selected from a class of compounds known as biopolymers.

<sup>84</sup>  
Claim 150 (amended). The composition of matter of claim 146 in which at least one of said distinct polyatomic parent molecular species is selected from the group comprising proteins, peptides polypeptides, carbohydrates, oligonucleotides and glycoproteins.

<sup>85</sup>  
Claim 151 (amended). The composition of matter of claim 146 in which at least one of said distinct polyatomic parent molecular species is not selected from the group comprising poly (ethylene glycol)s.

<sup>1186</sup>  
~~COX~~  
Claim 152 (amended). The composition of matter of claim 146 in which the mass spectrum of said distinct populations of multiply charged polyatomic ions comprises a coherent sequence of peaks in which the ions of each peak differ from the ions of immediately adjacent peaks by one adduct charge, the set of mass/charge (m/z) values for the ions of the peaks in said coherent sequence leading by appropriate calculations to a value for the molecular weight (Mr) of the distinct parent polyatomic molecular species from which are formed the polyatomic ions whose mass spectrum comprises said coherent sequence of peaks.

<sup>87</sup>  
Claim 153 (amended). The composition of matter of claim 146 in which at least one of said distinct polyatomic parent molecular species has a value for said molecular weight (Mr) not less than 5000.

<sup>88</sup>  
Claim 154 (amended). The composition of matter of claim 146 in which at least one of said distinct polyatomic parent molecular species has a value for said molecular weight (Mr) not less than 7000.

89

Claim 155 (amended). A composition of matter comprising one or more distinct populations of multiply charged polyatomic ions, each of said multiply charged polyatomic ions in said distinct populations being characterized by the symbol  $x_i$ , the numerical value of  $x_i$  being the  $m/z$  value of said one of said multiply charged ions such that  $x_i = Mr/i + ma$  wherein  $Mr$  is the molecular weight of a distinct parent polyatomic molecular species from which all of said multiply charged polyatomic ions in said distinct population of multiply charged polyatomic ions are derived,  $i$  is an integer equal to the number of adduct charges attached to a molecule of said distinct parent polyatomic molecular species to form one of said multiply charged polyatomic ions, and  $ma$  is the effective average mass (which can sometimes be negative) of each said adduct charge, each of said distinct populations of multiply charged polyatomic ions comprising a plurality of sub-populations, the ions of each sub-population having the same values for  $i$ ,  $ma$  and  $Mr$  and therefore the same value of  $x_i$ , said distinct populations of multiply charged polyatomic ions including at least one population in which all the ions have values of  $i$  greater than three.

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Claim 156 (amended). The composition of matter of claim 155 in which all the ions in said at least one population have values of  $i$  greater than six.

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Claim 157. The composition of matter of claim 155 in which at least one of said distinct polyatomic parent molecular species is selected from a class of compounds known as biopolymers.

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92 89  
Claim 158 (amended). The composition of matter of claim 155 such that the mass spectrum of the ions in at least one of said distinct populations of multiply charged Polyatomic ions, formed from at least one of said distinct polyatomic parent molecular species, comprises a sequence of peaks, one peak for each value of  $x_i$  found by mass analysis of the ions of said distinct population of multiply charged Polyatomic ions, the set of values for  $x_i$  including one for each value of  $i$  beginning with a smallest value of  $i$  <sup>not less than four</sup> and extending to a largest value not less than seven.

93 89  
Claim 159 (amended). The composition of matter of claim 155 in which at least one of said distinct polyatomic parent molecular species has a value of said molecular weight,  $Mr$ , not less than 5000.

94 89  
Claim 160 (amended). The composition of matter of claim 155 in which at least one of said distinct polyatomic parent molecular species has a value of said molecular weight,  $Mr$ , not less than 7000.

95 89  
Claim 161 (amended). A composition of matter comprising one or more distinct populations of multiply charged Polyatomic ions, each one of said multiply charged Polyatomic ions in said one or more distinct populations being characterized by the symbol  $x_i$ , the numerical value of  $x_i$  being the  $m/z$  value of said multiply charged polyatomic ion such that  $x_i = Mr/i + ma$  wherein  $Mr$  is the molecular weight of a distinct parent polyatomic molecular species from which all of said multiply charged Polyatomic ions in one of said distinct populations of multiply charged Polyatomic ions are derived,  $i$  is an integer equal

to the number of individual adduct charges attached to a molecule of said distinct parent polyatomic molecular species to form one of said multiply charged polyatomic ions,  $m_a$  is the effective average mass of said individual adduct charges (which can be negative), each of said distinct populations of ions comprising a plurality of sub-populations, the ions of each sub-population having the same values for  $i$ ,  $m_a$  and  $M_r$  and therefore the same value of  $x_i$ , the value of  $i$  being at least three for every ion in at least one of said distinct populations of multiply charged polyatomic ions, said composition of matter being formed by:

*(cont'd)*  
dispersing a solution containing said polyatomic parent molecular species into a bath gas as charged droplets, said dispersing taking place in the presence of an electric field, and

allowing the solvent of said solution to evaporate from said charged droplets until at least some molecules of said parent polyatomic parent molecular species become dispersed in said bath gas as said multiply charged ions.

Claim 162 (amended). The composition of matter of claim 161 in which every ion in at least one of said distinct populations of multiply charged polyatomic ions has a value of  $i$  not less than five.

Claim 163 (amended). The composition of matter of claim 161 in which every ion in at least one of said distinct populations of multiply charged polyatomic ions has a value of  $i$  not less than seven.

98  
Claim 164. The composition of matter of claim 161 in which at least one of said distinct polyatomic parent molecular species is selected from a class of compounds known as biopolymers.

99  
Claim 165 (amended). The composition of matter of claim 161 in which at least one of said distinct polyatomic parent molecular species is not selected from the group comprising poly (ethylene glycol)s.

160  
Claim 166 (amended). The composition of matter of claim 161 in which at least one of said distinct polyatomic parent molecular species has a value of said molecular weight, Mr, not less than 5000.

Claims 167-187 have been cancelled without prejudice.

161  
Claim 188. A composition of matter comprising one or more populations of polyatomic gaseous ions, at least one of said populations comprising multiply charged ions formed from the same chemically distinct parent species of polyatomic neutral molecules, said same chemically distinct species of polyatomic neutral molecules not including synthetic polymers such as poly (ethylene glycol)s, all of said multiply charged ions, formed from said same chemically distinct species of polyatomic neutral molecules, having at least three charges.

162  
Claim 189. A composition of matter according to Claim 188 in which all of said multiply charged polyatomic ions, formed from said chemically distinct species of polyatomic neutral molecules, have at least five charges.

Claim <sup>103</sup> ~~190~~ A composition of matter according to Claim <sup>101</sup> ~~188~~ in which all of said multiply charged polyatomic ions, formed from said chemically distinct species of polyatomic neutral molecules, have at least seven charges.

Claim <sup>104</sup> ~~191~~ A composition of matter comprising one or more populations of polyatomic gaseous ions, at least one of said populations of polyatomic ions comprising multiply charged ions formed from the same chemically distinct parent species of polyatomic neutral molecules, said chemically distinct parent species of polyatomic molecules not being selected from the class comprising oligomers of synthetic polymers such as poly (ethylene glycol)s, the number of charges on each ion defining the charge state number of that ion, said at least one of said populations of polyatomic multiply charged ions comprising a plurality of sub-populations, one such sub-population for each possible integral value of charge state number beginning with a smallest value not less than three and extending to a largest value not less than five.

*cont'd*

Claim <sup>105</sup> ~~192~~ A composition of matter according to Claim <sup>104</sup> ~~191~~ in which said smallest value of charge state number is not less than five and said largest value is not less than seven.

Claim <sup>106</sup> ~~193~~ A composition of matter according to Claim <sup>104</sup> ~~191~~ in which said smallest value of charge state number is not less than seven and said largest value is not less than ten.

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Claim 194. A composition of matter comprising one or more populations of polyatomic gaseous ions, all of the ions in at least one of said populations comprising multiply charged polyatomic ions having a net charge equal to or greater than three elementary charges and a composition characterized by the empirical chemical formula (Cc Hh Nn Oo Ss Pp Tt Uu Vv Ww Yy) wherein upper case letters C, H, N, O, S, P stand respectively for the elements Carbon, Hydrogen, Nitrogen, Oxygen, Sulfur, Phosphorous and T, U, V, W, Y each stand for other elements in the Periodic Table, the lower case subscript letters associated with each of said upper case letters symbolizing an integer equal to the number of atoms of the corresponding element in said ion, all the ions with three or more charges in at least one of said one or more populations of ions having compositions such that the number of different subscripts c, h, o, n, p, s, t, u, v, w, y having values greater than zero is five or less, said ions not being derived from a member of the class of synthetic polymers that includes poly (ethylene glycol)s.

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Claim 195. A composition of matter as in Claim 194 in which all the ions in said at least one population of multiply charged polyatomic ions have at least five charges.

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Claim 196. A composition of matter as in Claim 194 in which all the ions in said at least one population of multiply charged polyatomic ions have at least seven charges.

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Claim 197. A composition of matter comprising one or more populations of gaseous ions, at least one of said populations comprising multiply charged polyatomic ions having a net charge equal to or greater than three elementary charges and a composition characterized by the empirical chemical formula (Cc Hh Nn Oo Ss Pp Tt Uu Vv Ww Yy) wherein upper case letters C, H, N, O, S, P stand respectively for the elements Carbon, Hydrogen, Nitrogen, Oxygen, Sulfur, Phosphorous and T, U, V, W, Y each stand for other elements in the Periodic Table, the lower case letters symbolizing an integer equal to the number of atoms of the corresponding element in said ion, all the ions with three or more charges in at least one of said one or more populations of polyatomic ions having compositions such that the number of different subscripts c, h, n, o, p, s, t, u, v, w, y having values greater than zero is greater than five.

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Claim 198. A composition of matter as in Claim 197 in which all the ions in said at least one population of multiply charged polyatomic ions have at least five charges and a composition such that the number of different subscripts c, h, n, o, p, s, t, u, v, w, y having values greater than zero is greater than five.

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Claim 199. A composition of matter as in Claim 197 in which all the ions in said at least one population of multiply charged polyatomic ions have at least seven charges and a composition such that the number of different subscripts c, h, n, o, p, s, t, u, v, w having values greater than zero is greater than five.

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